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Fabrication of high-performance (Ba,K)Fe_2As_2 superconducting wires by powder-in-tube method

Q.P. Ding[a,b*], T. Prombood[a], S. Mohan[a], Y.Tsuchiya[a], Y. Nakajima[a,b], T. Tamegai[a,b]

[a]*Department of Applied Physics, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*
[b]*JST, Transformative Research-Project on Iron Pnictides (TRIP), 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*

Abstract

(Ba,K)Fe_2As_2 superconducting wires have been fabricated by *ex-situ* powder-in-tube method. In addition to the pure (Ba,K)Fe_2As_2 wires, silver powder was also used as a chemical addition to improve the performance of these superconducting wires. The transport critical current density (J_c) has reached 1.3×10^4 A/cm^2 at 4.2 K under self field in the wire with Ag addition. The self-field J_c is the highest among all the reported Fe-based superconducting wires so far. We have also performed magneto-optical imaging to this (Ba,K)Fe_2As_2 superconducting wire with Ag addition, and intragranular J_c of 6.0×10^4 A/cm^2 at 20 K is obtained, which is similar to the estimation from *M-H* measurement.

Keywords: (Ba,K)Fe_2As_2; powder-in-tube (PIT); superconducting wires; critical current density; magneto-optical imaging

1. Introduction

Iron-based superconductors have been investigated intensively during the past 3 years since the discovery of iron-pnictide $LaFeAsO_{1-x}F_x$ (1111) showing superconductivity ~ 26 K [1]. The studies were not only restricted to the underlying physics, but also towards the applications. The high upper critical field (H_{c2}) and critical current densities (J_c) make the iron-based superconductors very promising for practical applications [2]. Superconducting wires and tapes of 1111, (Ba,K)Fe_2As_2 (122), and Fe(Te,Se) (11) have been successfully fabricated [3-16]. Among these reported superconducting wires, the highest J_c in 11 is only ~ 1000 A/cm^2 [5], 4000 A/cm^2 in 1111 [11] and 1.0×10^4 A/cm^2 in 122 [12]. Hence 122 is the most promising among iron-based superconductors compared with 1111 and 11. For 1111, some fluorine will be lost after each sintering process, making it deviate from the optimal condition and producing more impurities. For 11, the transition temperature T_c, H_{c2} and even J_c are all much lower than 122. In this study, we have fabricated high-performance (Ba,K)Fe_2As_2 superconducting wires through *ex-situ* powder-in-tube (PIT) method. We investigated the effect of Ag addition on the superconducting properties of (Ba,K)Fe_2As_2 superconducting wires.

2. Experimental

$Ba_{0.6}K_{0.4}Fe_2As_2$ polycrystalline samples were synthesized by solid-state reaction method. Ba pieces (Kojundo Chemical Laboratory, 99%), K ingots (same as above, 99.5%), and FeAs powder were used as raw materials. FeAs

* Corresponding author. Tel.: +81-3-5841-6848 ; fax: +81-3-5841-6848 .
E-mail address: ding@ap.t.u-tokyo.ac.jp .

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was prepared by placing stoichiometric amounts of As pieces (Fukuzawa Electric, 99.99999%) and Fe powder (Kojundo Chemical Laboratory, 99%) in an evacuated quartz tube and reacting them at 1065 ^{o}C for 10 h after heating at 700 ^{o}C for 6 h. A mixture with a ratio of Ba: K: FeAs = 0.6: 0.44: 2 was placed in an alumina crucible. The whole assembly was sealed in a quartz tube, and slowly ramped up to 1100 ^{o}C in 20 h followed by cooling down to room temperature naturally. 10% K was added in order to compensate the reaction with quartz tube at high temperature. Powder-in-tube (PIT) method was used to fabricate $Ba_{0.6}K_{0.4}Fe_2As_2$ superconducting wires. As-prepared $Ba_{0.6}K_{0.4}Fe_2As_2$ polycrystalline sample was ground into fine powder with an agate mortar and pestle in a nitrogen-filled glove box. The powder was divided into two parts. One part was filled into Ag tube with OD 4.5 mm and ID 3 mm (The Nilaco Corpoartion, 99%) directly. 15% in weight of Ag powder (Kojundo Chemical Laboratory, 99%) was mixed with the other part and then put into Ag tube. The assembly was cold drawn into a square wire with diagonal dimension of about 0.6 mm. The as-drawn wire was cut into shorter wires with length ~ 40 mm, and then sealed in an evacuated quartz tube. The sealed wires were put into a muffle furnace and heated up to 700^{o}C with a ramping rate of 100 ^{o}C/h, and kept at this temperature for 24 h. Then the furnace was switched off and cooled to room temperature naturally.

The phase identification of the sample was carried out by means of powder X-ray diffraction (M18XHF, MAC Science) with Cu-*Kα* radiation generated at 40kV and 200 mA. Bulk magnetization is measured by a superconducting quantum interference device SQUID magnetometer (MPMS-5XL, Quantum Design). Current-voltage (*I*-*V*) measurements were performed by four-probe method with silver paste for contact. *I*-*V* measurements were performed in a bath-type cryostat (Spectromag, Oxford Instruments). For local magnetic characterization, we applied magneto-optical (MO) imaging. For this purpose, the wire was cut using a wire saw, and the cross sections were polished with a lapping film. A Bi-substituted iron-garnet indicator film is placed in direct contact with the sample, and the whole assembly is attached to the cold finger of a He-flow cryostat (Microstat-HR, Oxford Instruments). MO images are acquired by using a cooled CCD camera with 12-bit resolution (ORCA-ER, Hamamatsu). To enhance the visibility of the local magnetic induction and eliminate the signals from the impurity phases, a differential imaging technique is employed [17, 18].

3. Results and discussions

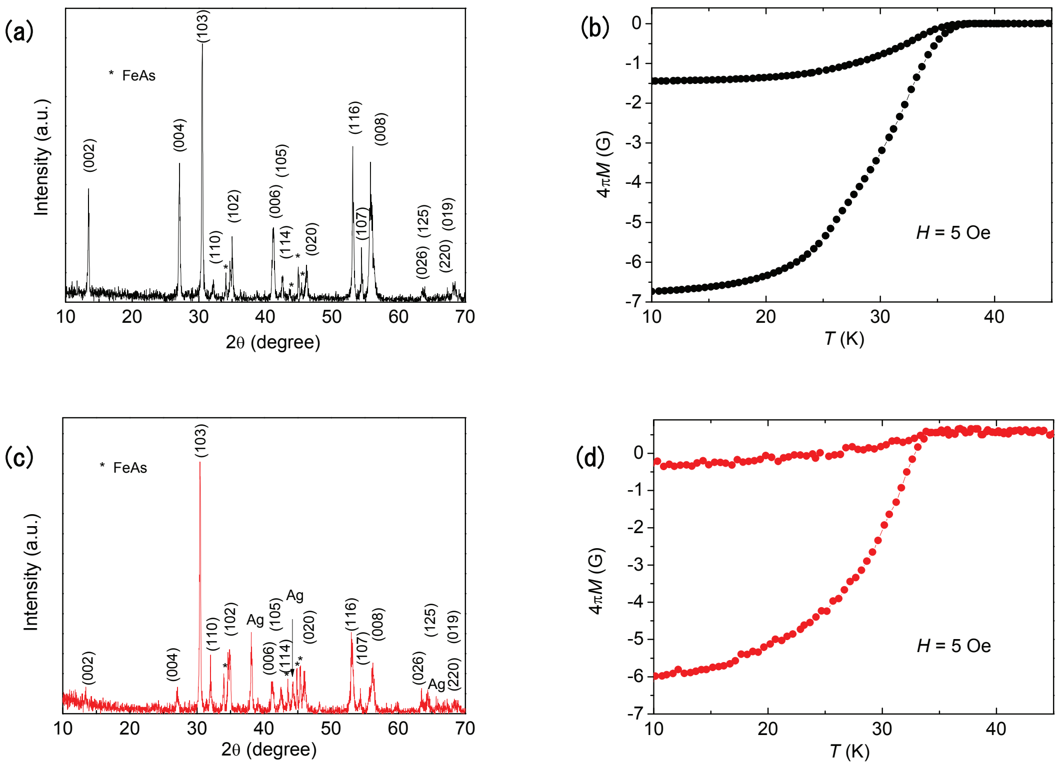

Figure 1. Powder X-ray diffraction pattern of (a) $Ba_{0.6}K_{0.4}Fe_2As_2$ polycrystalline sample and (c) $Ba_{0.6}K_{0.4}Fe_2As_2$ PIT superconducting wire with Ag addition. Temperature dependence of magnetization of (b) $Ba_{0.6}K_{0.4}Fe_2As_2$ polycrystalline sample and (d) $Ba_{0.6}K_{0.4}Fe_2As_2$ PIT superconducting wire with Ag addition measured at 5 Oe.

Figure 1(a) shows the X-ray diffraction pattern of as-prepared polycrystalline sample. All the peaks were well indexed using a space group of I4/mmm except for small peaks as an impurity phase marked by asterisks. The compound crystallizes in a tetragonal structure and the impurity phase is identified as remaining FeAs. Temperature dependences of zero-field-cooled (ZFC) and field-cooled (FC) magnetization at 5 Oe of the $Ba_{0.6}K_{0.4}Fe_2As_2$ polycrystalline sample are shown in Fig. 1(b). The sample shows an onset of diamagnetism at around 38 K. This value is similar to the optimally-doped $(Ba,K)Fe_2As_2$. Figure 1(c) shows the X-ray pattern of the core of the sintered wire with Ag addition. Compared with the polycrystalline sample before filling into the Ag tube, several new peaks which were assigned to the added Ag powder were detected. Shown in Fig. 1(d) are the temperature dependences of magnetization of the heated wire with Ag addition. The transition temperature T_c has decreased to 35 K after sintering.

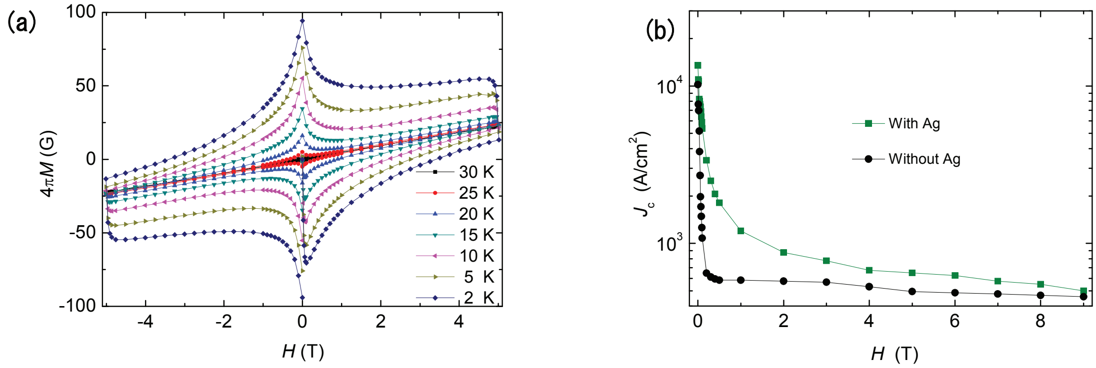

Figure 2 (a) Magnetic field dependences of magnetization of $Ba_{0.6}K_{0.4}Fe_2As_2$ PIT superconducting wire with Ag addition; (b) Field dependences of the transport J_c's in $Ba_{0.6}K_{0.4}Fe_2As_2$ PIT superconducting wires with and without Ag addition.

Magnetic hysteresis curves of the wire with Ag addition are shown in Fig. 2(a). By using the Bean model with an assumption of field–independent J_c, intragranular critical current density J_c^{intra} for polycrystalline samples can be evaluated from the magnetization hysteresis loops [19]. According to the Bean model, J_c^{intra}[A/cm^2] is given by $J_c^{intra} = 30\ \Delta M/d$, with an assumption that intergranular critical current is zero, where ΔM[emu/cc] is M_{down} - M_{up}, M_{up} and M_{down} are the magnetization when sweeping field up and down, respectively, d[cm] is the average diameter of the grains in the polycrystalline sample. From the MO image in Fig. 3, a typical grain size is ~ 10 μm. J_c^{intra} estimated from M-H curve is ~ 6.0 $\times 10^4$ A/cm^2 at 20 K under zero field. Above 20 K, conventional low-temperature superconductors cannot be used. 122 is a good candidate for applications in this temperature range.

We investigated the transport J_c of those superconducting wires with and without Ag addition. In superconductors, driving force competes with pinning force. If the former wins over the latter, measurable voltages could be observed. Here we adopt E = 1 μV/cm as a criterion to define transport J_c. Transport J_c as a function of field at 4.2 K for both wires with and without Ag addition are shown in Fig. 2(b). Compared with the wire without Ag addition, the transport J_c was enhanced by Ag addition. The self-field transport J_c is 1.3$\times 10^4$ A/cm^2, and 1.0$\times 10^4$ A/cm^2 at 4.2 K for the wire with and without Ag addition, respectively. Not only the self-field J_c, but the strong field-dependence feature has also been improved remarkably through Ag addition, especially at low fields (below 2 T).

Figures 3(a)-3(c) depict MO images of the transverse cross section of the Ag-added wire in the remanent state after applying a 500 Oe field for 0.25 seconds which was subsequently reduced down to zero at temperatures between 50 K and 30 K. In these images, the bright regions correspond to the trapped flux in the sample. These images are similar to the MO images of 1111 and 11 polycrystals and tapes [20-24]. At all temperatures, bright intensities are restricted in small regions, implying that the intergranular current density is much smaller compared with the intragranular current density. The intragranular current density decreases gradually as the temperature is increased towards T_c. Figure 3(d) shows the magnetic induction profiles along the dashed line in figure 3(a). From the magnetic induction profile we calculated the intragranular critical current density. In this calculation, we roughly estimate the intragranular critical current density by $J_c^{intra} \sim dB/dx$. The estimated J_c^{intra} is ~ 6.0 $\times 10^4$ A/cm^2 at 20 K for typical grains. This value is consistent with the value estimated from the M-H curve.

4. Conclusion

In conclusion, X-ray diffraction, magnetization, resistivity, transport critical current density and magneto-optical measurements were performed on $Ba_{0.6}K_{0.4}Fe_2As_2$ superconducting wires with and without Ag addition prepared through PIT method. A transport critical current density of 1.3×10^4 A/cm^2 at 4.2 K under self field is obtained in a wire with Ag addition. The self-field J_c value is the highest value among reported Fe-based superconducting wires so far. The intragranular J_c estimated from MO imags is consistent with the value calculated from the *M-H* curve. Addition of two or more elements to improve the self-field J_c and reduce its field dependence, and fabricating denser wires by applying pressure is the subject of ongoing research.

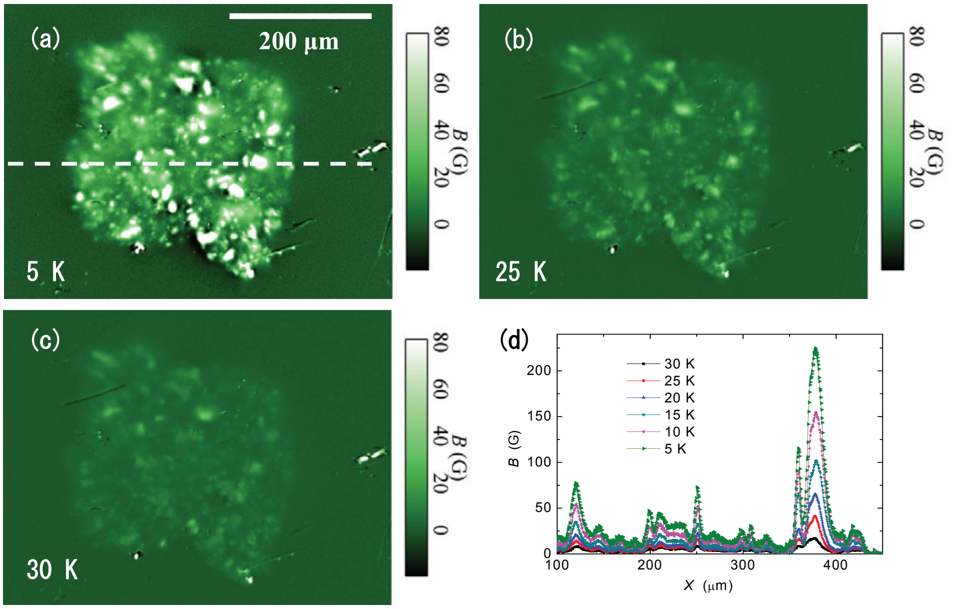

Figure 3. MO images in the remanent state of the $Ba_{0.6}K_{0.4}Fe_2As_2$ PIT superconducting wires tape at (a) 5 K, (b) 25 K, and (c) 30 K after cycling the field up to 500 Oe for 0.25 seconds; (d) The local magnetic induction profiles at different temperatures taken along the dashed lines in (a).

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